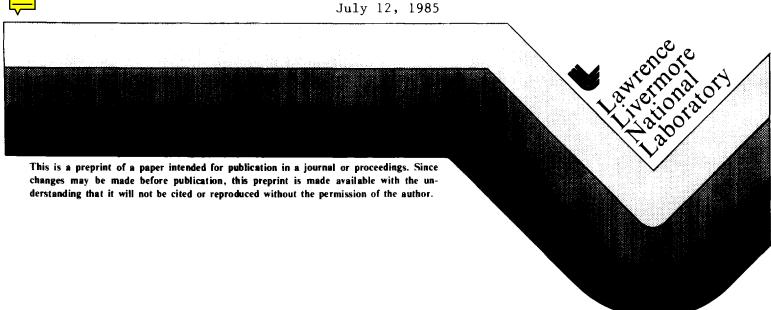
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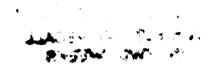
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CONDENSED MATTER AT HIGH SHOCK PRESSURES

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CONDENSED MATTER AT HIGH SHOCK PRESSURES

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Experimental techniques are described for shock waves in liquids: Hugoniot equation-of-state, shock temperature and emission spectroscopy, electrical conductivity, and Raman spectroscopy. Experimental data are reviewed and presented in terms of phenomena that occur at high densities and temperatures in shocked He, Ar, N2, C0, SiO2-aerogel, H2O, and C6H6. The superconducting properties of Nb metal shocked to 100 GPa (1 Mbar) and recovered intact are discussed in terms of prospects for synthesizing novel, metastable materials. Ultrahigh pressure data for Cu is reviewed in the range 0.3-6TPa (3-60 Mbar).

I. INTRODUCTION

Significant advances in shocked, condensed matter have been made in recent years through diagnostic development, specimen fabrication, and theory. The nature of this work is summarized in the proceedings of recent Topical Conferences on Shock Waves in Condensed Matter.^{1,2} General areas of concentration have been the properties of fluids and solids at high densities, temperatures, and strain rates and the nature of materials processed and recovered from high dynamic pressures. In this paper the main emphasis will be on dense fluids at shock pressures up to 100 GPa (1 Mbar), compressions up to 5-fold over initial liquid density, and shock temperatures up to 10,000K. At such extreme conditions several phenomena occur including molecular repulsion, rotation, vibration, and dissociation, electronic and molecular ionization, and chemical decomposition and reaction. Examples from the literature will be reviewed and new results will be presented to illustrate these phenomena.

The properties of dense molecular fluids at high pressures and temperatures have three principal applications: the equation of state and chemistry of high explosives, the nature of the interiors of the outer planets (Jupiter, Saturn, Uranus, and Neptune), and hydrodynamic applications. The relation between these areas and shock-compression studies was discussed previously.³

II. EXPERIMENTS

The experiments are performed using shock waves generated by the impact of planar projectiles onto planar target specimens. While, historically, strong shock waves have been generated using high explosives, 4 in recent years most research has utilized impactors launched by light-gas guns. 5 Maximum impact velocities are 2 and 8 km/s for single-stage and two-stage light-gas guns, respectively. A Ta impactor at 2 and 8 km/s achieves shock pressures in Al of 30 and 200 GPa, respectively. Gas guns have several advantages over explosive shock generators. Shock strength is simply tuneable by choice of impactor material and velocity, shock-wave profile is simply tuneable by choice of impactor thickness and design, shock pressures twice as large as with plane wave explosive systems are available, and facilities are operated in a conventional laboratory setting. The current state of temporal

and spatial resolution for shock-wave diagnostics means that gun facilities can now be scaled down substantially without compromising the ability to perform novel and diverse research with a well characterized and simply tuneable shock driver.

The experiments discussed here were performed using a two-stage light-gas gun described previously. $^{6-8}$ The two driving media or stages are gunpowder reaction products and H₂ gas. The gunpowder drives a heavy piston which compresses the H₂ gas. When the H₂ pressure reaches $^{\sim}$ 100 MPa (1 kbar) a rupture valve opens and the compressed gas accelerates a light ($^{\sim}$ 15g) projectile to velocities up to 8 km/s along a smooth bore launch tube. Strong shock waves are generated by impact of projectile onto target positioned about 0.5m beyond the muzzle of the launch tube. Projectile velocity is measured in free flight between muzzle and target by a flash radiograph system. A more detailed description of two-stage gun operation and shock-wave diagnostics is available.

The fundamental measurement is equation of state which determines the pressure, density, and specific internal energy achieved in shock compression experiments. These experiments are based on the Rankine-Hugoniot relations and shock impedance matching. The measured quantities are impactor velocity, shock velocity, and initial density. The method is described elsewhere for single- and double-shocked liquids. 10,11 Having measured the equation of state, other properties can be measured to study the nature of the state; for example, shock temperature, electrical conductivity, and Raman spectra. In these experiments the impactor velocity is also measured and the state achieved is obtained by shock impedance matching using the previously determined equations of state. Figures 1a and 1b illustrate experimental configurations for single and double-shock Hugoniot experiments. In the former, shock velocity is measured in the liquid, and in the latter it is measured in an anvil behind the liquid.

A shock temperature or emission spectrum measurement in a transparent liquid is illustrated in Fig. lc. Radiation emitted from the shock front exits the unshocked portion of the specimen and is directed to either a 6-channel time-resolved pyrometer 12 , 13 or to a spectrograph and 1000 -channel, time-integrated, gated, linear diode array. 14 For simple dense fluids like Ar, both molecular dynamics and continuum mechanics calculations indicate that the shock front is about 10 Å and $<10^{-12}\rm s$ thick. 15 , 16 Thus, for simple shocked fluids the emission spectra are fit to graybody or blackbody spectra and the assumption is made that the temperature so derived is characteristic of equilibrated shocked material emitting light from behind the shock front through an optically thin shock front. For more complex fluids, for example those undergoing chemical reactions, the shock front can be optically thick, so that the equilibrium temperature is not observed while nonequilibrium spectral features might be.

A two-probe electrical conductivity experiment for a shocked liquid is illustrated in Fig. ld. A current is established in the shunt resistor, and the time-resolved voltage across the electrodes in parallel with the shunt resistor is measured as the shock transits the electrodes. The cell resistance versus known electrical conductivities is calibrated in separate experiments prior to the shock experiment.

The microscopic nature of shocked fluids can be investigated by pulsed Raman spectroscopy, which measures the distribution of molecular vibrational frequencies. The experimental arrangement is illustrated in Fig. 2. Light from a 10 ns 0.2J KrF (284 nm) laser pulse is incident normally on a specimen of shocked liquid. Light from spontaneous Raman scattering, 10^{-10} J total, is viewed at 45° off axis, dispersed by a spectrograph, and recorded on a gated, linear diode array. The laser light is focussed several mm behind the shock front and so this technique requires that the shocked specimen remain transparent.

In the case of transparent, brittle, solid specimens, like silica aerogel, the configuration for an equation-of-state experiment in which shock velocity is measured is shown in Fig. 3. 19 Because the shock front radiates strongly in the visible, the stepped rear surface is coated with $\sim 1000~\text{Å}$ of Au to absorb thermal radiation until shock breakout at the coated surface. By temporally streaking the optical emission from the coated, stepped surface, the transit time of the shock across the step height can be measured. This technique is similar to that used previously to measure transit times for laser-driven shock waves in metals. 20 The shock temperature is measured by using a simple, uncoated specimen disc and sending light emitted from the shock front to a pyrometer or spectrograph.

Several additional measurements are made in shocked solids, including time-resolved interface velocity measurements by optical interferometry, 21 , 22 the generation of dynamic, planar, isentropic compression waves by graded-density impactors, 23 the detection of phase transitions by both the measurement of longitudinal sound speeds 24 and shock temperatures, 25 and flash X-ray diffraction from crystals in the shock state. 26 , 27

In addition to the diagnostic developments major advances have resulted from specimen fabrication. For example, in order to investigate the high-density regime of molecular fluids, higher-density liquid specimens were shocked, rather than gases. In order to liquify most small molecules at atmospheric pressures, cryogenic temperatures are required. Thus, each specimen holder was a small cryostat 10,28,29 - a different one for each experiment. States of high-temperature matter in expansion relative to crystal density have been studied because of the recent availability of fine-grained, transparent, porous SiO₂ aerogel having \sim 100 Å pore size. 19 The fine pore size means that the material is uniform and equilibrium will be achieved much faster than the time scale of the experiments. However, this material is extremely brittle and requires vacuum holders and diamond cutting tools to achieve the required micron surface flatness. Dynamic isentropic compression is now possible because of the development of impactors with linearly varying density (shock impedance) produced by careful sedimentation of metallic powders in an epoxy-like matrix.²³ These examples illustrate the importance of special materials and fabrication techniques in this field.

III. THERMAL EQUILIBRIUM

Shocked condensed matter is generally in thermal equilibrium during the measurements owing to the high densities and temperatures. For example, liquid N2 shocked to 30 GPa¹⁰ has a density of 2 g/cm³ (4 x 10^{22} N2/cm³) and a shock temperature of ~ 6000 K.³0 Assuming the mean free path between collisions is a molecular diameter and that molecular velocity is thermal, then the intermolecular collision time is $\sim 10^{-13}$ s. The N2 molecular vibration time is $\sim 10^{-14}$ s. The shock front width or equilibration time in shocked dense Ar is $\lesssim 10^{-12}$ s. If All these times are much less than the time resolutions of the diagnostic systems which are $\sim 10^{-9}$ s and the duration of the experiments which are 10^{-7} - 10^{-6} s. Of course, phenomena which require longer than the experimental lifetime could produce deviations from thermal equilibrium. At the conditions considered here, however, examples of nonequilibrium behavior are difficult to identify unambiguously and are most likely to occur in chemically reacting systems.

IV. RESULTS

Equation-of-state experiments have been performed in the last few years for most small molecules including He, 29 H₂ and D₂, 28 N₂, 10 , 31 O₂, 10 air, Ar, 10 , 32 Xe, 33 CO, 11 CH₄, 11 C₆H₆, 34 n-C₄H₈ (polybutene), 34

 NH_3 17 and $H_20.17$ The maximum pressures and densities, which our group has achieved in these shocked liquids, are listed in Table I. The corresponding calculated and, in some cases, measured maximum temperatures are in the range 5,000-30,000 K.

Shock temperature measurements have been performed for a number of these fluids: $H_2O_1^{13}$ NH3,35 Ar,36 N2,36 C6H6,34 and n-C4H8.34 Temperature is an important thermodynamic parameter because it provides information about the distribution of internal energy between thermal and internal degrees of freedom. In the case of C6H6 and possibly n-C4H8 the radiating temperatures from the shock fronts are substantially lower than the temperatures calculated theoretically assuming chemical equilibrium. This difference suggests that the radiating temperatures are not representative of shocked matter in thermal and chemical equilibrium behind the shock front.34

The electrical conductivity has been measured for shocked H_20 , 17 , 37 NH3, 38 N2, 39 O2, 39 and C_6H_6 . 39 The purpose is to determine the magnitude and mechanisms of electrical conduction and in so doing to possibly obtain information about the nature of the fluids which cannot be obtained from state variables. For example, the high conductivity of shocked H_20 indicates a significant amount of H_20 molecules are chemically ionized above 20 GPa shock pressure. 37 This proposal motivated the Raman spectroscopy measurements described below in order to look for ionic species like OH- and H_30^+ . The electrical conductivities of shocked N_2 and O_2 are even higher than for H_20 and electrons are probably the dominant conduction mechanism.

The spontaneous Raman spectra of shocked water has been measured from 7-26 GPa¹⁸ to try to identify molecular and ionic species from spectral features. At 26 GPa water is compressed two-fold and heated to 1700 K. The stimulated Raman scattering from shocked benzene has been measured at 1.2 GPa, indicating the existence of the molecular state at this shock pressure. ⁴⁰ The emission spectrum of benzene has been measured to 59 GPa to try to identify chemical species in or close to the shock front. ¹⁴

V. DISCUSSION

Intermolecular repulsion is the dominant effect at high densities and temperatures. The rare gases are the simplest materials to study in this respect, because the physical properties are determined solely by the repulsive potential, provided electronic excitation is negligible. The He Hugoniot data are interpreted only in terms of an effective pair potential, the calculated shock temperatures as large as 20,000 K being much smaller than the ionization potential.²⁹

For Ar the upper limit of the "cold" range is a single-shock pressure of 36 GPa, a compression of 2-fold over initial liquid density, and a calculated temperature of 12,000 K. The pair potential derived from Ar shock-wave data is in excellent agreement with pair potentials derived from molecular-beam scattering data, indicating that two-body interactions dominate in the dense fluid. The pair potential derived from Ar shock-wave data has the exponential-six form: 41

$$\phi(r) = \varepsilon \left\{ \left[\frac{6}{\alpha - 6} \right] \exp \left[\alpha (1 - r/r^*) \right] - \left[\frac{\alpha}{\alpha - 6} \right] \left(\frac{r^*}{r} \right)^6 \right\} , \qquad (1)$$

where r is the intermolecular distance. The Ar Hugoniot data are shown in Fig. 4. Curve D was calculated using only the potential of Eq. (1) and agrees with the data below 36 GPa. At higher pressures the data fall well below Curve D. Agreement with the data is achieved by taking into account electronic excitation and the density dependence of the electron energy gap.³² A similar result was obtained for Xe.³³ A fundamental conclusion of the rare gas study is that high shock temperature is a very useful probe of the electronic structure at high density and pressure.

The shock-compression data for non-polar molecules can be calculated using the potential of Eq. (1) scaled via corresponding states and by taking into account molecular vibrational and rotational degrees of freedom. The energy parameter ε is scaled on critical temperature and the distance parameter r* on the cube root of critical volume. The procedure is valid systematically to a compression of 2.5 times liquid density for N2, C0, Xe, O2, CH4, and CO2.⁴¹

The Hugoniot data for liquid $N_2^{10,31,42}$ and CO are shown in Fig. 5. No and CO are isoelectronic, they have virtually identical critical points, and in these experiments they had the same initial density, 0.81 g/cm³. Thus, they are expected to have the same Hugoniot curve by the arguments in the preceding paragraph. The curve through the data up to 15 and 30 GPa for CO and N2, respectively, was calculated using a potential scaled from Ar via corresponding states⁴ and is in excellent agreement with both the N₂ and CO data. This agreement is evidence that No and CO retain their molecular structure up to 30 and 15 GPa, respectively. Deviations from molecular behavior are interpreted as decomposition in both. In the CO case a large number of product molecules are probably formed by chemical reaction. Nitrogen probably undergoes a dissociative phase transition. In fact, double-shock points for nitrogen starting from 20-25 GPa lie above the principal Hugoniot in pressure-volume space. This is the only known material in which this has been observed and is evidence that these points are in a phase transition region. Comparison with theory indicates that the shock temperature is driving the diatomic to monatomic transition in nitrogen at lower pressures than required at room temperature. Above 70 GPa the two Hugoniot curves merge again and atom density appears to dominate over the chemical nature.

Hugoniot data for silica aerogel are shown in Fig. 6. This aerogel is initially 20-fold expanded relative to α -quartz density. Although the specimens were shock-compressed 6 to 7-fold, the material is still 3-fold expanded relative to crystal density. In an experiment shocked to 6.7 GPa a shock temperature of 10,800 K was measured. Thus, these experiments access a new regime for accurate laboratory measurements on high-temperature expanded-volume states of glass. This material has several important potential uses. It can be a very low shock-impedance equation-of-state standard. It can be used to generate and study few-eV inertially-confined plasmas, as for shocked gases. Its equation of state may be used to understand cratering phenomena because SiO2 is a major geological material.

Water is a complicated fluid in that the H₂O molecule is polar and the intermolecular potential is not spherically symmetric. Intermolecular H-bonds have a strong influence on the structure of water, especially at low pressures. The electrical conductivity of shocked water is quite large, about 20 (ohm-cm)⁻¹ from 30-60 GPa.¹⁷ This conductivity has been interpreted in terms of ionization: $2 \text{ H}_2\text{O} \rightarrow 0\text{H}^- + \text{H}_3\text{O}^+$. The high conductivity indicates that significant, if not total, ionization does occur.

In order to identify the chemical species in shocked water, spontaneous Raman spectroscopy experiments were performed in the shock pressure range 7.5-26 GPa. Only a broad OH-stretch band is observed at all pressures. A spectrum for 11.7 GPa is shown in Fig. 7. No definite indications of OH- or $\rm H_3O^+$ ions are observed based on the positions of their vibrational bands at ambient. The position of the OH-stretch band is virtually independent of shock pressure and the shape of the OH-stretch band changes in such a way as to indicate the destruction of intermolecular H-bonds, as would be expected. These bonds have a characteristic energy of $^{\sim}$ 1000 K and the shock temperature is about 1,700 K at 26 GPa. The analysis seems to indicate that the $\rm H_2O$ molecule remains the dominant scattering component with increasing pressure. The $\rm H_3O^+$ ion is not observed because its presence is associated

with hydrogen-bonded H_2O complexes which are destroyed at these conditions. Absence of H_3O^+ means that OH^- and H^+ are most probably the conduction mechanism in strongly shocked water. Evidently, the hydroxyl ion cannot be distinguished from H_2O at high shock pressures.

Chemical decomposition and reaction occurs in shocked hydrocarbons. Benzene, for example, has the distinct volume collapse of a phase transition in its Hugoniot curve at 13 GPa. 44 Optical absorption experiments show that benzene retains its molecular nature up to 13 GPa. 45 The volume collapse at 13 GPa has been interpreted by the formation of dense carbon phases. 34,46 The emission spectra from shock fronts in benzene have been measured in the range 22-59 GPa. 14 A spectrum at 50 GPa is shown in Fig. 8. The spectrum has a broad graybody thermal component with nonequilibrium features superimposed. Features in the spectral range are coincident with the Swann bands for the C2 molecule and provide the first spectral evidence for the formation of carbon phases in shocked hydrocarbons.

High dynamic pressure is also used to synthesize new materials. For example, stoichiometric Nb₃Si with a high superconducting transition temperature $T_c = 18 \text{ K}$ has only been synthesized by processing a nonsuperconducting phase at 100 GPa (1 Mbar) shock pressures.47 develop the experimental and computational techniques to process materials at 100 GPa shock pressures and recover them for investigations of material structure and physical properties, Nb metal was recently recovered intact from peak dynamic pressures in the range 60-120 GPa.48 Pressure and effective-plastic-strain histories were calculated using a finite element hydrodynamic computer code. Recovered specimens were characterized by X-ray diffraction, metallography, Vicker's hardness, and $T_{\rm C}$. The maximum change observed in a T_C of 9.18 K for the unshocked specimen was 0.035 K in the specimen shocked to a maximum pressure of 0.6 Mbar. Shock processing V₃Si at 100 GPa depresses its $T_{\rm C}$ by 1.8 K from the initial value of 16.4 K. 49 These results indicate that shock-induced defects and disorder have a relatively weak effect on the T_C of equilibrium phases and suggest that shock-synthesized metastable superconductors might have T_C's close to intrinsic values for the ordered material.

Finally, shock-impedance-match experiments have been performed at ultrahigh pressures using two-stage guns to accelerate impactors to 8 km/s, chemical explosives to drive impactors to 15 km/s, and strong shock waves generated in proximity to underground nuclear explosions. 50-56 At ultrahigh pressures nuclear-explosive-driven experiments are the only ones that provide the required accuracy because of the long shock transit times and steady profile of the shock wave. The Hugoniot data for Cu is summarized in Fig. 9, which is a plot of shock velocity versus mass velocity. The data in this figure span the shock pressure range 0.3-6 TPa (3-60 Mbar). At lower pressures the data fit a linear shock velocity-mass velocity relation, shown as the solid line, which extends up to 1.5 TPa. At higher pressures the data deviates systemmatically below the extrapolation of the linear fit, which is probably indicative of a transition to Thomas-Fermi behavior at pressures above 10 TPa. The smooth nature of the data are striking considering the variety of techniques that were employed over the \sim 25 years in which this data was accumulated.

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Table 1. Summary of shock-compression experiments for fluids. T_0 is initial temperature, ρ_0 is initial density, ρ_{max} is maximum density, and P_{max} is maximum shock pressure.

Liquid	т _о (К)	(g/cm ³)	^p max (g/cm³)	P _{max} (GPa)a
Helium	4	0.12	0.68	56
Deuterium	20	0.17	0.96	76
Hydrogen	20	0.07	0.23	10
Nitrogen	77	0.81	2.8	82
0xygen	77	1.2	3 .3	86
Carbon Monoxide	77	0.81	3.0	69
Methane	111	0.42	1.6	92
Benzene	295	0.89	2.4	70
Polybutene	295	0.89	2.7	210
Argon	87	1.4	3.8	91
Xenon	165	3.0	9.6	130
Ammonia	230	0.69	1.8	64
Water	295	1.0	3.5	220

al GPa = 10 kbar.

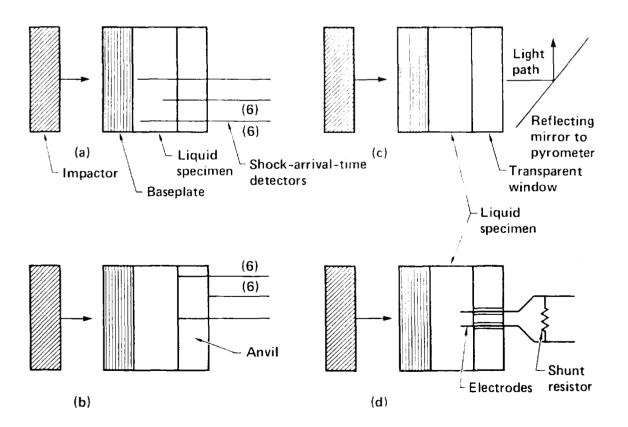


Figure 1. a) Schematic of single-shock equation-of-state experiment.10 b) Double-shock equation-of-state experiment.11 c) Shock temperature13 or emission spectroscopy14 for transparent liquid specimen. d) Electrical conductivity experiment.17

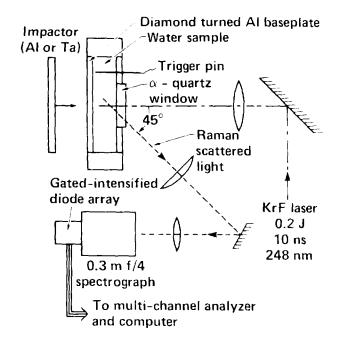


Figure 2. Schematic of pulsed, spontaneous, Raman scattering experiment in shocked water. 18

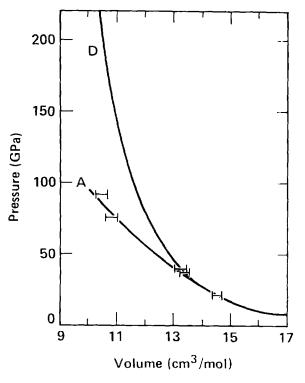


Figure 4. Hugoniot data for shocked liquid Ar.10 Curve A was calculated taking into account interatomic repulsion and electronic excitation across volume-dependent electron energy gap. Curve D was calculated neglecting electronic excitation. 32

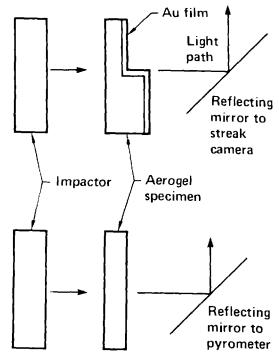


Figure 3. Schematic of equation-of-state (top) and shock temperature (bottom) experiments for shocked transparent silica aerogel. At top, Au coating screens thermal radiation from a streaking camera until shockwave reaches rear surfaces of stepped target. At bottom, light emitted from the shock front is sent to a pyrometer during transit time of shock through specimen. 19

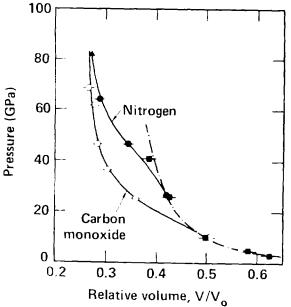


Figure 5. Hugoniot data for shocked liquid N_2 10,31,42 and isoelectronic CO.11 Dash-dot curve is theory⁴¹ for both molecular N_2 and CO. Initial volume of both is 34.7 cm³/mole.

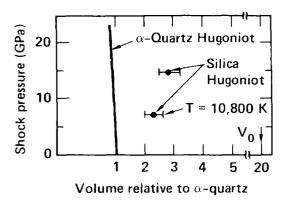


Figure 6. Hugoniot data for silica aerogel, which is porous SiO_2 expanded 20-fold relative to α -quartz density. ¹⁹ Although aerogel is compressed substantially, the final densities are still less than crystal density. Indicated temperature was measured pyrometrically.

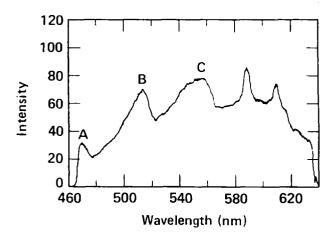


Figure 8. Emission spectrum from benzene shocked to 50 GPa. 14 Features labelled A, B, C are coincident with the Swann bands for C₂ molecule.

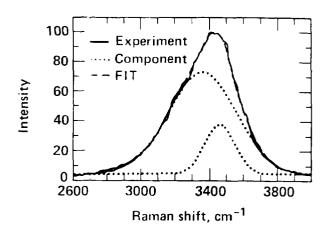


Figure 7. Spontaneous Raman spectrum from water shocked to 11.7 GPal8 using system in Fig. 2. Spectrum is sum of two gaussian components shown.

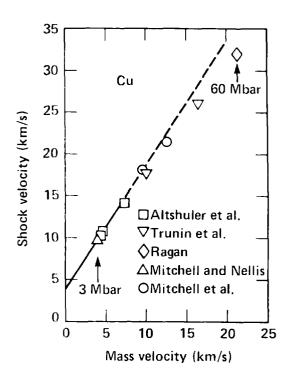


Figure 9. Shock velocity versus mass velocity for Cu spanning range 0.3-6 TPa (3-60 Mbar).50-56